

POTENTIAL USE COCONUT MILK AS ALTERNATIVE TO ALKALI SOLUTION FOR GEOPOLYMER PRODUCTION

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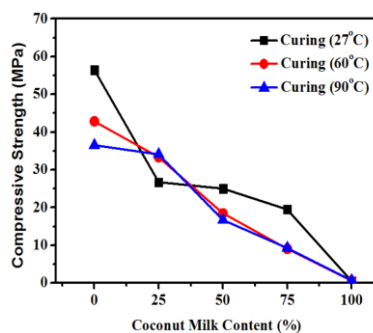
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Graphical abstract



Abstract

This work aims to verify the feasibility of utilizing coconut milk as the alkali activator solution in geopolymer production and the impact on mortar properties; geopolymer mortar is still more expensive than ordinary Portland cement mortar simply because the cost of alkali solution. Coconut milk is extensively available in Malaysia and very rich in potassium and sodium. In this research, the coconut milk was used as alkali solution (100%) at first, and then replaced by NaOH, Na₂SiO₃ and in the last stage mixed with NaOH and Na₂SiO₃ at 50%. Normal solution component of Na₂SiO₃ and NaOH with 8 M, and used as control samples. Binder to fine aggregate (B:A) and solution to binder (S:B) ratios were fixed at 1.5 and 0.30 respectively. Multi blend binder based geopolymer mortar are used in this study. The samples were cured with different conditions, cured at room temperature and oven temperature of 60 and 90°C. Compressive strength tests were carried out to determine the properties of hardened mortar. The samples prepared with coconut milk showed low compressive strength as compared to control samples. The results demonstrated that using coconut milk as alternative to alkali solution in geopolymer industry is not a viable option.

Keywords: Geopolymer mortar, alkali activator solution, coconut milk, compressive strength

Abstrak

Kajian ini bertujuan untuk mengesahkan kemungkinan menggunakan santan kelapa sebagai larutan pengaktif alkali pada sifat-sifat mortar geopolimer; mortar geopolimer adalah lebih mahal daripada Portland simen mortar kerana kos larutan pengaktif alkali masih sangat mahal. Santan kelapa yang terdapat dengan meluasnya di Malaysia mempunyai kandungan kalium dan natrium yang tinggi. Dalam kajian ini, santan kelapa telah digunakan sebagai larutan alkali (100%) pada mulanya, dan kemudian digantikan dengan NaOH, Na₂SiO₃ dan diperingkat akhir dicampur dengan NaOH and Na₂SiO₃ pada tahap 50%. Komponen larutan normal Na₂SiO₃ and NaOH dengan 8 M, telah digunakan sebagai sampel kawalan. Binder kepada nisbah agregat halus dan natrium silikat kepada natrium hidroksida telah ditetapkan pada 1.5 dan 3.0. Pelbagai campuran binder berasaskan mortar geopolimer telah digunakan dalam kajian ini. Dalam kajian, sampel telah diuji dalam keadaan suhu yang berbeza iaitu pada suhu bilik dan suhu oven pada tahap 60°C dan 90°C. Ujian kekuatan mampatan telah dijalankan untuk menentukan sifat-sifat kekerasan mortar. Sampel yang disediakan dengan santan kelapa menunjukkan kekuatan mampatan yang rendah jika dibandingkan dengan dengan sampel kawalan. Keputusan menunjukkan bahawa menggunakan santan kelapa sebagai alternatif kepada larutan alkali dalam industri geopolimer bukan merupakan pilihan yang baik.

Kata kunci: Penyembuhan diri sendiri, halatuju denyutan ultrasonik and ujian kemampuan

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1.0 INTRODUCTION

Every year, more than billion tons of greenhouse gas emissions were added by the global cement industry, and contributed around 7% of the total emissions to the earth's atmosphere. Attempts by researchers are beginning to reduce emissions of carbon dioxide (CO₂) for a cleaner environment. The use of wastes materials from industry such as palm oil fuel ash, fly ash, metakaolin and furnace slag to substitute the amount of ordinary Portland cement (OPC) in concrete is one of many options which was widely studied [2-5]. Geopolymer being a polymer of inorganic alumina-silicate are synthesized from the alkaline activation of different types of aluminosilicate materials, is one of the alternatives to fabricate environmentally safe concrete, that can contribute to reducing CO₂ emission. In the early of 1979s, Joseph Davidovites introduced the geopolymerization technology as a more environment-friendly process [6]. Materials that are rich in silicon and aluminium of geological origin or wastes from industrial are utilized to synthesize the geopolymer concrete [7-8]. Alkaline silicate or/and hydroxides are used as activation of alumina-silicate materials at ambient or oven curing regime [9].

In the 1940s, Purdon started studying the feasibility use sodium hydroxide as alkali-activator solution to activate the furnace slag [10]. Alkali activated system which contained calcium silicate hydrate (CSH) and aluminosilicate phases was invented in the late 1950s by Glukhovsky [11]. Two types of geopolymer binding systems which are silica-calcium (Si + Ca) with a mild alkaline solution and silica-aluminium (Si + Al) with medium to high alkaline solution [12]. For CASH binding system, the source materials are class F fly ash and metakaolin due to silica and alumina content as the main composition. Meantime, for the CSH system, furnace slag (GBFS) was included due to its main composition which is silica and calcium. The hydration products of these two systems are also different, whereas in the (Si + Ca) system, calcium silicate hydrate (CSH) is the main product, while zeolite like polymers is the main products for CASH system [13].

In geopolymerization, alkaline solution plays an important role. The most common alkaline solution used in geopolymerization is a combination of sodium hydroxide (NaOH) or potassium hydroxide (KOH) and sodium silicate (Na₂SiO₃) or potassium silicate (K₂SiO₃). In this study, a combination of sodium hydroxide and sodium silicate was chosen as the alkaline liquid. Sodium based solutions were chosen because they are cheaper than potassium based solutions. Generally sodium hydroxide and sodium silicate are readily available in the market in the form of pellets and gel (liquid) [14]. The most commonly used alkaline activator is NaOH [15-19].

Research has shown the combination of sodium hydroxide and sodium silicate as alkaline activator solution increased geopolymerisation percentages as

compared to using solely hydroxide [12]. In the process of geopolymerisation, Xu and Van Deventer [20] has found that additional silica (Si) is needed in various sources of alumina-silicate mineral materials during the production of geopolymer. Alkali hydroxide is required for the dissolution process of aluminosilicate sources, while Na₂SiO₃ solution acts as binder [11].

One of the disadvantages of geopolymer mortar is that it is still more expensive than OPC mortar. Alkali activator solution contributed more than 60% of the geopolymer mortar cost. Studies have to be made in the area of manufacturing process of sodium hydroxide so as to make it less expensive [21].

Coconut milk is extensively available in Malaysia and it is rich in potassium and sodium ions 60% and 25% respectively [22]. Hence, the present paper is based on an attempt to use coconut milk as an alternative of alkali activator solution with. The mechanical properties of geopolymer mortar were studied in detail.

2.0 METHODOLOGY

2.1 Materials

The FA (class F) being the source of aluminosilicate is obtained from the Tanjung Piai power station (Malaysia), which is further used for making the GPMs. Granulated blast furnace slag (GBFS) and palm oil, fuel ash (POFA) were used in the experiments. OPC was used for making OPC mortar to compare the compressive strengths of geopolymer mortars. The chemical compositions of OPC, Fly ash, GBFS and POFA are given in Table 1. Multi blended binder was used to prepare the geopolymer mortar with 2.95 (Si/Al) and 1.08 (Ca/Si) ratio as shown in table 2. River sand with 2.36 mm maximum size as fine aggregate was used in this experiment. Mineral water was used in all the experiments. The chemical composition of coconut milk used in this study is presented in table 3. The alkali activators used were of sodium hydroxide and sodium silicate solution as control sample and other sixth patches components (Table 4).

2.2 Preparation of Alkalis

Sodium hydroxides solution (8M) was prepared and left for 24 h before mixing with sodium silicate. Sodium hydroxide mixtures and sodium silicate solutions were left for 1 day before using them in geopolymerization process. Other patches of solution prepared by using coconut milk with various components are given in Table 4.

Table 1 Chemical composition (XRF) test

Item.	OPC	GBFS	FA	POFA
SiO ₂	20.80	30.80	57.20	64.20
Al ₂ O ₃	4.70	10.9	28.80	4.25
Fe ₂ O ₃	3.40	0.64	3.67	3.13
CaO	65.30	51.80	5.16	10.20
MgO	1.50	4.57	1.48	5.90
K ₂ O	0.40	0.36	0.94	8.64
Na ₂ O	0.10	0.45	-	-
SO ₃	2.70	-	-	-
LOI	0.90	0.22	-	1.73

Table 2 (Si/Al) and (Ca/Si) ratio of blend components

MIX	SiO ₂	Al ₂ O ₃	CaO	Si/Al	Ca/Si
%	37.80	12.82	41.07	2.95	1.08

Table 3 Chemical composite of coconut milk

Materials	CaO	Fe ₂ O ₃	MgO	K ₂ O	Na ₂ O	Al ₂ O ₃
(%)	5.60	0.01	5.88	58.82	24.71	0.01

2.3 Mix Proportion of Geopolymer Concrete

The geopolymer mortar was formulated using the customary method as OPC mortar, due to the equal proportionate density of OPC mortar (2240 kg/m³) [15]. In the present mix design of geopolymer mortar, multi binder to fine aggregates was fixed at 1.5 for all mixtures. The ratio of sodium silicate (NS) to sodium hydroxide (NH) solution was maintained at 3.0. 8 M NaOH solutions. Seven mixtures were formulated. The measurement/extent of fine aggregates and multi binder were remained constant while alkali activator was partly replaced by coconut milk, as given in Table 4. In this study, Portland cement as the control mixture was formulated according to ASTM C109 with the purpose of comparing the geopolymer mortars. This detailed mix design of geopolymer mortar mixtures can be seen in Table 4.

2.4 Casting of Geopolymer Mortar Mixes

The conventional techniques used for OPC mortar were adopted. OPC mortar was prepared using the traditional method whereby the multi-binder was mixed together for 2 min initially. Next, saturated surface dry (SSD) fine aggregate was combined to the binder and mixed for about 3 min. During the process, various alkali solutions were added to the dry materials and the mixing continued for 2 min. For each of the ready mixtures, 27 cubes were cast in a 50 mm steel model in two layers, and then compacted for 20 second using a vibration machine. The casted samples were left molded in the laboratory at room temperature.

2.5 Curing of Geopolymer Mortar

Nine samples were left to cure at room temperature after de-moulding as shown in Figure 1a. The other 18 samples were transferred to the oven with two patches for heat curing at 60°C and 90°C for 24 hours as shown in Figure 1b. The samples were then left at room temperature after curing until the day of testing.

**Figure 1** Curing method (a) ambient curing (b) oven curing

2.6 Tests

According ASTM C109, compressive strength test was conducted to evaluate the strength of samples as depicted in Figure 2. Samples were tested after 3, 7 and 28 days and the results were reported based on the average of three samples.

**Figure 2** Compressive strength test

Table 4 Geopolymer mortar mixtures with coconut milk

MIX	Binder (%)				B/A	S/B	Alkali activator solution (%)					
	GBFS	FA	POFA	H.L			NS	NH	CO.M	NS/NH	M	W
M1	74	15	10	1	1.5	0.30	75	25	-	3.0	8	-
MP1	74	15	10	1	1.5	0.30	-	-	100	-	-	-
MP2	74	15	10	1	1.5	0.30	-	25	75	-	8	-
MP3	74	15	10	1	1.5	0.30	75	-	25	-	-	-
MP4	74	15	10	1	1.5	0.30	37.5	12.5	50	3.0	8	-
MP5	74	15	10	1	1.5	0.30	75	-	-	-	-	25
MP6	74	15	10	1	1.5	0.30	100	-	-	-	-	-

3.0 RESULTS AND DISCUSSION

3.1 Analysis of Compressive Strength

Geopolymer mortars strength developed with age during different temperature curing periods is presented in Figures 3 to 8. Compressive strength after 3, 7 and 28 days and different temperature curing conditions was studied.

Geopolymer mortar strength with different components of the solution and curing temperatures are shown in Figure 3 to 5. Figure 3. Shows early compressive strength after 72 hours. Mixture based sodium silicate as solution with 100% (MP6) showed high strength early on (38.22, 52.44 and 54.08) at 27, 60 and 90°C respectively. Coconut milk as alkali solution 100% (MP1) did not show any results; coconut milk used as an alternative to sodium silicate (MP2) and mixed with sodium hydroxide in the ratio of 3:1 showed low strength 10MPa for curing temperatures. Geopolymer mortar activated with 50% coconut milk and 50% solution prepared by mixed sodium silicate with sodium hydroxide (MP4) also showed lower than 6Mpa compressive strength. Solution prepared by mixing sodium silicate and coconut milk with 3.0 ratios (MP3) showed results also less than 10 MPa at room temperature. However, strength more than 19.80 Mpa was noted at oven curing temperatures. Mixture (MP5) prepared by using sodium silicate mixed with water in 3:1 ratio, demonstrated strength as 18.54, 28.14 and 28.02 MPa with different curing temperature. In Figure 3 mixtures (MP1, MP2, MP3 and MP4) depicted strength less than a mixture (MP5). The compressive strength of geopolymer mortar increased when the amount of Na_2SiO_3 increased. Moreover, the use of Na_2SiO_3 helped to improve the geopolymerisation process by accelerating the dissolution of source material.

The strength of aging geopolymer mortar after different curing temperature periods of 7 and 28 days are shown in Figure 4 and 5. Geopolymer mortar compressive strength prepared with coconut milk (MP1-4) was still less than the strength of control sample (M1), also the results still less than the strength of geopolymer prepared with sodium silicate solution in mixture MP5 and MP6.

The compressive strength of different mixtures cured at room temperature is displayed in Figure 6. Geopolymer mortar prepared with coconut milk

showed weak results for different age specimens. However, geopolymer mortar prepared with normal solution (NS+NH) showed the best results and achieved compressive strength 58.44 MPa after 28 days.

Compressive strength of samples cured in oven at 60 and 90°C is recorded in Figure 7 and 8. Geopolymer mortar activated with coconut milk still showed less strength than mixtures prepared without coconut milk. Geopolymer mortar activated with sodium silicate (MP5 and MP6) showed higher compressive strength than normal solution prepared with sodium silicate and sodium hydroxide.

Coconut milk is rich of Na_2O and K_2O as clearly display in Table 3. The fact has increased Na_2O content improve the compressive strength of geopolymer mortar by increase the dissolution of silicate and aluminium as reported by Vickers, L. *et al.* 2015 [23]. In addition to the $\text{SiO}_2\text{:Na}_2\text{O}$ ratio, water to binder ratio needs to be considered. Higher water contents reduce the pH (lower OH^- concentration) and reduce the rate of dissolution.

The PH of normal alkali solution (sodium hydroxide and sodium silicate) higher than 13 and that lead to high concentration of OH^- and increase the dissolution and achieved high strength as shown in mixture (M1). The PH of coconut milk is 7 [24] and that mean the concentration of OH^- is very low in the activating solution leads to low strength (0.46, 19.8, 26.71 and 25.01 MPa) geopolymers (MP1-4) even if Na_2O levels are high.

The addition of sodium silicate to the activating solution enhances the polymerization process of the ionic species present in the system as shown in GPM mixtures (M1 and MP6). Activating solutions made from sodium hydroxide and sodium silicate needs to be optimised in terms of not only the $\text{SiO}_2\text{:Na}_2\text{O}$ ratio but also the actual amounts. Equilibrium between NaOH and Sodium silicate in the solution should be reached in order to maintain the system with a high pH and a high level of soluble silica as a display of GPM mixture (M1), that explain clearly the effective normal solution to achieve higher strength at ambient curing temperature compared with other mixtures (MP1-6).

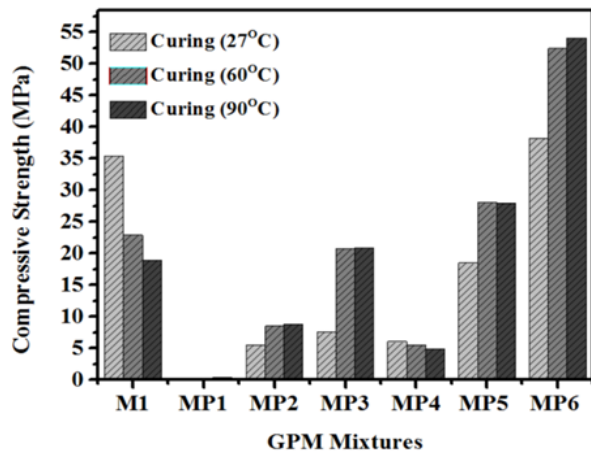


Figure 3 Development compressive strength with varies curing temperatures after 3 day

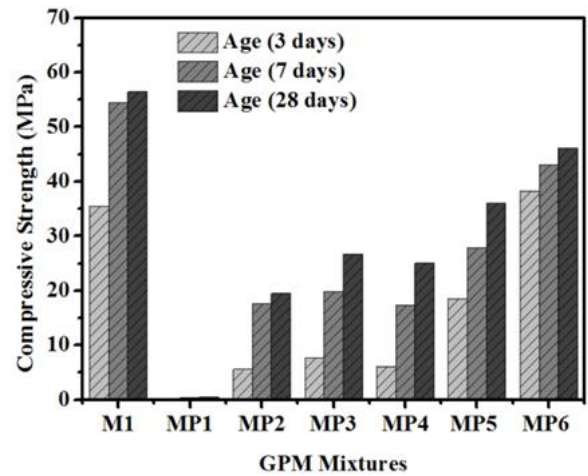


Figure 6 Development compressive strength with time curing (27°C)

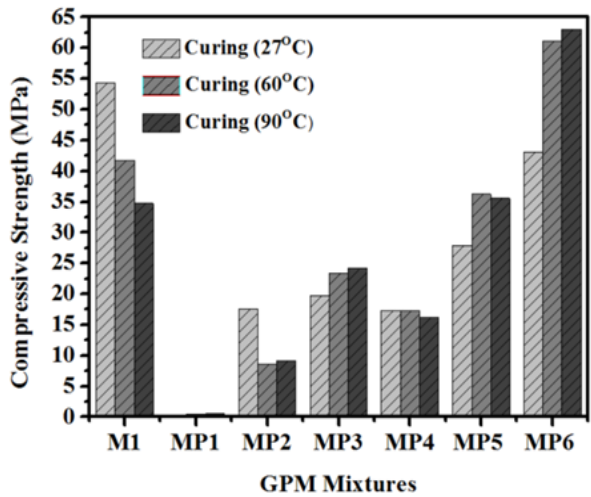


Figure 4 Development compressive strength with varies curing temperatures after 7 day

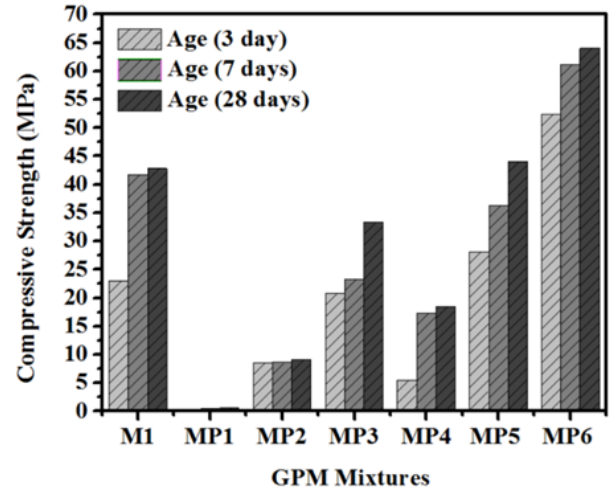


Figure 7 Development compressive strength with time curing (60°C)

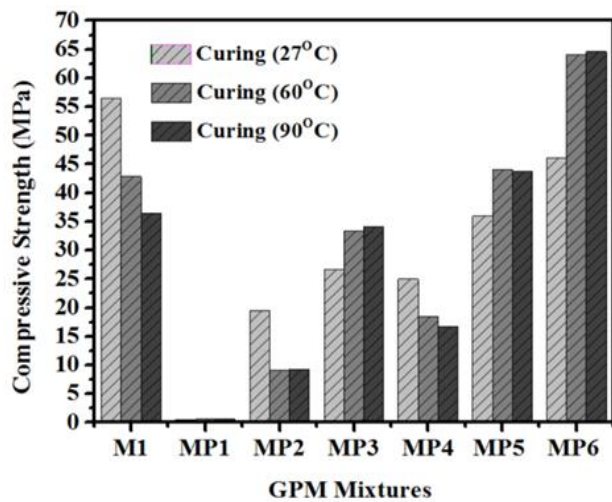


Figure 5 Development compressive strength with varies curing temperatures after 28 day

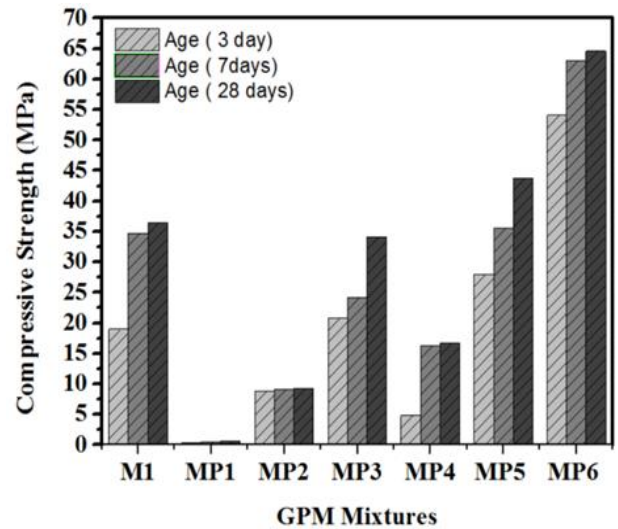


Figure 8 Development compressive strength with time curing (90°C)

Figure 9 display the impact coconut milk content in alkali solution, evidently the increase coconut milk content lead to reduce the compressive strength from 58.44 Mpa to 0.48 Mpa with 0 and 100% of coconut milk content respectively. When the activating solution is a mixture of sodium hydroxide and coconut milk the $\text{SiO}_2/\text{Na}_2\text{O}$ and the OH^- concentration is reduced which increase the coconut milk content. This involves a reduction in pH leading to lower amounts of aluminium and silicon dissolved from the multi blend [25]. This could explain the low strengths obtained with this activator solution.

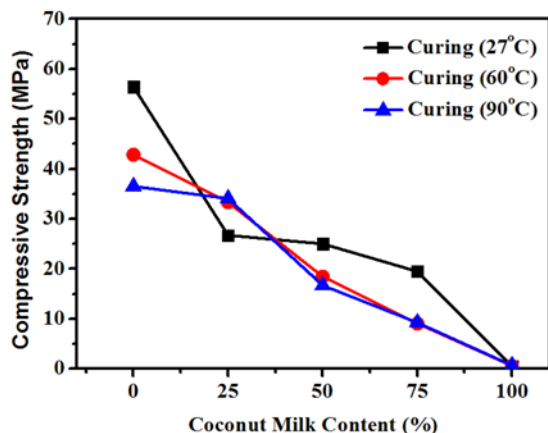


Figure 9 Development compressive strength of GPM with different coconut milk content after 28 days.

The results of compressive strength of geopolymer mortar prepared with coconut milk (MP3) with content 25% as display in Table 4 above and cured at ambient temperature was depicted in Figure 8. The results of strength after 28 days still lower than conventional mortar (OPC). Also the results of GPM content coconut milk show lower results than geopolymer mortar prepared without coconut milk (MP5-6) for same reason Previously mentioned above the increase coconut milk reduce the concentration of OH^- and lead to reduce PH of solution from 13 to 7 and effect in dissolution of silicate.

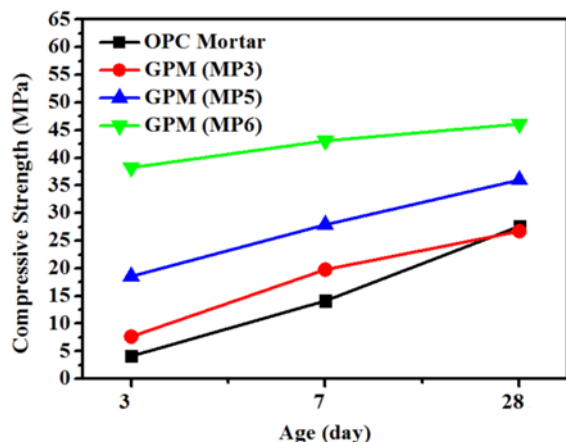


Figure 10 Compare the developed compressive strength of GPM with conventional mortar (OPC)

4.0 CONCLUSIONS

The following conclusions can be drawn from this preliminary study:

- Compressive strength results clearly indicated that the use of coconut milk cannot activate geopolymer binder as an alkali solution.
- Sodium silicates without sodium hydroxide solution could be suitable for high early compressive strength of the preparation geopolymer mortar.
- Sodium silicate used as alkali solution showed better performance in oven curing as compared with normal solution prepared from sodium silicate with sodium hydroxide.
- Solution prepared from sodium silicate and sodium hydroxide appeared to be more suitable for ambient curing condition.
- Increase coconut milk leads to reduction of OH^- concentration and the solution PH, that which impact on the dissolution of silicate and reduces the compressive strength.
- Increase coconut milk content leads to reduction of the $\text{SiO}_2/\text{Na}_2\text{O}$ and effect on the development of the compressive strength.

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Notations

GPM	Geopolymer mortar
NS	Sodium silicate (Na_2SiO_3)
NH	Sodium hydroxide (NaOH)
M	Molarity of sodium hydroxide

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